

RPR-9476

21-2-140

FEDERAL REPUBLIC OF GERMANY  
GERMAN PATENT OFFICE

(51) Int. Cl.<sup>2</sup>: F 25 j 1-00

**Offenlegungsschrift 24 40 215**

(21) File number: P 24 40 215.1

(22) Application date: 8/22/74

(43) Date of opening for public inspection: 3/4/76

(30) Convention priority:

(32) (33) (31) --

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(54) Title: **Process for liquefaction and undercooling of a low-boiling gas**

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(H 823)

H 74/048

La/p

8/21/74

### **Process for liquefaction and undercooling of a low-boiling gas**

The invention pertains to a process for liquefaction and undercooling of a low-boiling gas with coolants carried in several cooling loops.

A process has already become known for liquefaction and undercooling of natural gas, in which the natural gas is cooled in heat exchange with a propane loop and liquefied and undercooled in heat exchange with a mixed loop. Within the propane loop, propane is compressed, liquefied, and expanded in three steps. After each expansion step part of the propane is evaporated against natural gas and the multicomponent mixture of the mixed loop, as a result of which the natural gas is precooled and the multicomponent mixture, which consists essentially of nitrogen, methane, ethane, and propane, is partially condensed. Then the partially condensed multicomponent mixture is subjected to phase separation. The liquid fraction obtained in this process is undercooled, expanded, and evaporated against the liquefying natural gas, against the gaseous fraction obtained during phase separation, which likewise liquefies during this heat exchange, and against itself, as a result of which undercooling takes place. The liquefied gaseous fraction is likewise undercooled, expanded, and evaporated against natural gas and itself, as a result of which the natural gas and the liquefied gaseous fraction are undercooled. After evaporation, the two fractions are again jointly conveyed to the loop compressor of the mixed loop. (DT-OS 1,960,301.)

This known process is unfavorable from the energy point of view, since in particular the heat conversion in the mixed loop is very large. The total amount of cold required for undercooling or for liquefaction and undercooling of the fractions obtained during the phase-separation, in addition to the cold required for liquefying and undercooling the natural gas, must be supplied through the system.

Furthermore, the evaporation of the two fractions must take place at relatively low pressures, resulting in the fact that the suction pressure of the loop compressor is low and thus its effective compression work is high. Furthermore, it has been found that because a liquid mixture is evaporated in the coldest heat exchangers, the pressure drop in these heat exchangers is disadvantageously high.

The apparatus cost for performing the known process is also very high, since in the precooling range at least three expansion steps with a corresponding number of expansion valves and heat exchangers are required to achieve a somewhat good approximation of the heating curve of the propane to the cooling curve of the natural gas.

The invention is based on the goal of developing a simple and energy-advantageous process for liquefying and undercooling a low-boiling gas.

This goal is accomplished through the fact that the cooling and at least partial liquefaction of the gas take place in heat exchange with a liquid multicomponent mixture, and the complete liquefaction and undercooling of the gas take place in heat exchange with an expanded gaseous coolant.

The process in accordance with the invention is highly advantageous from both the apparatus and the energy viewpoint. As a result of the use of a mixed cooling loop for precooling and at least partial liquefaction of the gas, with the aid of a single loop at constant pressure, cold can be released over a broad temperature difference. This is not the case in the known processes. Here, for covering a similar temperature difference, several different pressure steps and correspondingly also several expansion valves and heat exchangers are required. Furthermore, in the case of the object of the invention, in the range of cooling and liquefaction of the low-boiling gas the evaporation of the liquid multicomponent mixture takes place not at constant but at sliding temperatures. Corresponding to the boiling diagram of the multicomponent mixture in this process, a certain mixture composition is assigned to each evaporation temperature. By appropriately selecting the course of evaporation and the composition of the multicomponent mixture, the temperature range of the evaporation can therefore be adapted very well to the cooling curve of the low-boiling gas. The temperature differences in the heat exchangers are small, and therefore the energy losses are slight.

The deep cooling of the gas, thus its complete liquefaction and undercooling, takes place according to the invention in heat exchange with a gaseous coolant which, according to a further feature, is advantageous cooled in heat exchange with the multicomponent mixture, and then expanded with

performance of work. Through this measure, even in the lowest temperature range a very good adaptation of the heating curve of the coolant to the cooling curve of the gas can be achieved, since both curves follow a relatively shallow path in this range and thus low temperature differences exist even in the coldest range. An additional advantage lies in the fact that the heat turnover of such an expander loop is relatively low, since the total amount of cold generated by the expansion can be transferred directly to the gas to be treated. It is not necessary, as in the known case, to use part of the cold generated in the loop for liquefying and undercooling the circulating medium itself. Furthermore, it has been found that in contrast to the known process, the pressure losses in the heat exchangers in which the initial heating of the expanded gaseous coolant takes place are very low. Furthermore, the expansion of the gaseous coolant can take place to a relatively high final pressure, which once again has an advantageous effect on the required compression work of the circulating compressor.

The process in accordance with the invention is especially suitable for the liquefaction of natural gas in so-called "base-load units."

In this case advantageously a multicomponent mixture is used, which is made up of hydrocarbons with one, two, three, or four, and possibly even more carbon atoms, whereas as the gaseous coolant nitrogen, thus a gas that boils at a lower point than natural gas, comes under consideration.

Additional explanations for the invention can be taken from the exemplified embodiments shown schematically in the figures, wherein the same device parts are respectively represented with the same reference numbers.

The figures show the following:

Figure 1, an embodiment of the invention for the liquefaction of natural gas

Figure 2, an additional embodiment

Figure 3, a third embodiment.

According to Figure 1, in which an exemplified embodiment of the invention for liquefaction and undercooling of natural gas is shown, the natural gas to be treated is introduced to the unit under a pressure of about 50 ata over a pipeline 1. After it has been cooled and completely liquefied in the heat exchangers 2 and 3, the natural gas is undercooled in a heat exchanger 4 and expanded over a valve 5 into a storage container 6. The flash gas obtained during the expansion is heated in the heat exchangers 4, 3, and 2 against natural gas and withdrawn as fuel gas from the unit over a pipeline 7.

The cold required for the cooling and liquefaction of the natural gas is supplied by a mixed loop, and the cold required for undercooling by a nitrogen expander loop.

In the mixed loop a multicomponent mixture consisting of methane, ethane, propane, and butane is compressed in the circulating compressor 8, partially condensed in the water cooler 9, and subjected to phase separation in the separator 10. The liquid fraction obtained in the separator 10 is expanded in valve 11 and evaporated in heat exchanger 2 against cooled natural gas, against the nitrogen of the expander loop and against the gaseous fraction obtained in the separator 10, which liquefies in this heat exchanger. The gaseous fraction obtained in the separator 10, after its liquefaction in the heat exchanger 2, is undercooled against itself in the heat exchanger 3, expanded in an expansion valve 12, and evaporated in the heat exchanger 3 against liquefying natural gas, against the nitrogen of the expander loop, and against itself. After further heating in the heat exchanger 2 it is conveyed back to the circulating compressor 8 together with the liquid fraction from the separator 10.

Within the nitrogen expander loop, the nitrogen compressed to the required final pressure in the brake blower 13 and cooled in the water cooler 14, is first cooled in the heat exchangers 2 and 3 against the multicomponent mixture, and then in an additional heat exchanger 15, against itself. Then the nitrogen is expanded in a work-performing manner in a turbine 16 and heated in the heat exchanger 4 against natural gas, which undercools during this heat exchange. The energy liberated by the work-performing expansion in the turbine 16 is transferred directly to the brake blower 13 coupled with the turbine 16. In the heat exchanger 15, the work-performing, expanded nitrogen is further heated in heat exchange with compressed nitrogen and conveyed to the first step 17 of a two-step circulation compressor 18 and then compressed to a moderate pressure. Then the nitrogen is again cooled, first in the water cooler 19 and then in the heat exchanger 2 against the multicomponent mixture. Then further compression of the nitrogen takes place in the second compression step of the circulating compressor 18. After another cooling in the water cooler 21, the nitrogen is now conveyed for final compression in the brake blower 13. The intermediate cooling of the nitrogen in the heat exchanger 2 after its compression in the first compression step 17 proves highly advantageous, since because of the volume flux reduced by the cooling, the energy demand of the second compression step 20 can be considerably reduced and this [compression step 20] can also be designed to be smaller.

It has proven particularly favorable from the energy viewpoint to already completely liquefy the



natural gas in heat exchange with the multicomponent mixture in the heat exchangers 2 and 3 and to use the nitrogen expansion loop exclusively for undercooling the natural gas.

An additional exemplified embodiment of the invention is shown in Figure 2. As in the exemplified embodiment according to Figure 1, the cooling and liquefaction of the natural gas takes place in the heat exchangers 2 and 3 in heat exchange with a mixed loop and the undercooling in the heat exchanger 4 in heat exchange with a nitrogen expansion loop, wherein now, however, the natural gas is so strongly undercooled in the heat exchanger 4 that no further flash gas is obtained during the subsequent expansion in valve 5. In contrast to Figure 1 furthermore, in this exemplified embodiment the compression of the nitrogen takes place in the loop compressor 18 in a single step.

However, one important difference between the two embodiments lies in the concept of the mixed loop.

According to Figure 2, the compression of the multicomponent mixture takes place in two steps in the compression steps 22 and 23. After the first compression step 23, the multicomponent mixture is partially condensed under moderate pressure in the water cooler 24 and subjected to a phase separation in the separator 25. The liquid fraction obtained in this process is undercooled in the heat exchanger 2 against itself, expanded in the valve 26, and then evaporated and initially heated in the heat exchanger 2 against the nitrogen of the expansion loop, against natural gas, against the gaseous fraction from the separator 25, and against itself. The gaseous fraction obtained in the separator 25 is compressed in the second compression step 23 to the final pressure of the loop, cooled in the water cooler 27, and liquefied in the heat exchanger 2. Then this fraction is undercooled in the heat exchanger 3, expanded in the valve 28, and evaporated in the heat exchanger 23 against the nitrogen expander loop, against liquefying natural gas, and against itself. The two fractions together are now again conveyed to the first compression step 22 of the circulation compressor. The concept described gives the following advantages: through the partial condensation and phase separation of the multicomponent mixture even after the first compression step, thus at intermediate pressure, the multicomponent mixture can be enriched to an increased extent with higher boiling hydrocarbons such as propane, butane, and possibly even higher boiling components, which has an advantageous effect on the cooling performance of the loop because of the relatively high heats of evaporation of these higher boiling hydrocarbons. It has been found that such an increase in the concentration of higher hydrocarbons in the multicomponent mixture is not possible without further effort.

in a mixed loop according to Figure 1. Here, in the case of a concentration increase, higher hydrocarbons are carried along into the lowest temperature range of the loop as a result of which the evaporation temperature in this range was undesirably increased, and solids deposited may cause fogging in the corresponding heat exchanger cross-sections. As a result of the partial condensation and separation of the higher boiling hydrocarbons even after the intermediate compression, on the other hand, the partial pressure of these hydrocarbons in the multicomponent mixture entering the lower temperature ranges is kept low enough so that no undesirable increase in the evaporation temperature takes place there.

An additional exemplified embodiment of the invention is shown in Figure 3.

According to this figure, the cooling and liquefaction of the natural gas drawn in over pipeline 1 takes place in the heat exchangers 2, 3, and 29 against evaporating multicomponent mixture, and the undercooling in the heat exchanger 4 takes place against work-performing, expanded nitrogen. The nitrogen expansion loop very closely resembles that shown in Figure 2. However, here the lowest cooling of the nitrogen takes place before its expansion in the turbine 16 in heat exchange with the multicomponent mixture, which evaporates in the heat exchanger 29.

The mixed loop according to Figure 3 differs from that described in Figure 2 essentially in that the gaseous fraction obtained in the separator 25 during the intermediate condensation of the multicomponent mixture is not directly led into the second compression step 23, but is again partially condensed in the heat exchanger 2 and subjected in separator 30 to an additional phase separation. The liquid fraction obtained in this process is undercooled in the heat exchanger 3 and expanded in the valve 31, while the gaseous fraction obtained in the separator 30 is now compressed to the circulation pressure in the second compression step 23. This fraction is liquefied and undercooled in the heat exchangers 2, 3, and 29, expanded in valve 32, and evaporated in the heat exchanger 29 against the nitrogen of the expander loop, against natural gas, and against itself.

### Claims

1. Process for liquefaction and undercooling of a low-boiling gas by cooling with coolants conveyed in several cooling loops, characterized in that the cooling and at least partial liquefaction of the gas takes place in heat exchange with a liquid multicomponent mixture, and the complete liquefaction and undercooling of the gas takes place in heat exchange with an expanded gaseous coolant.
2. Process in accordance with Claim 1, characterized in that the gaseous coolant, after it is compressed, is cooled in heat exchange with the multicomponent mixture and then expanded in a work-performing manner.
3. Process in accordance with Claims 1 and 2, characterized in that the compression of the gaseous coolant takes place in several steps and that at least after a compression step the gaseous coolant is cooled in heat exchange with the multicomponent mixture.
4. Process in accordance with Claims 1 to 3, characterized in that as the gaseous coolant a gas is used, the boiling point of which is lower than the boiling point of the gas to be liquefied.
5. Process in accordance with Claims 1 to 4, characterized in that the low-boiling gas to be liquefied is natural gas and the gaseous coolant is nitrogen.
6. Process in accordance with Claims 1 to 5, characterized in that the low-boiling gas to be liquefied is completely liquefied in heat exchange with the multicomponent mixture.
7. Process in accordance with Claims 1 to 6, characterized in that the multicomponent mixture, after its at least one-step compression, is partially condensed in at least one condensation step, and that the gaseous fraction obtained in each condensation step is separated and liquefied in heat exchange with the expanded liquid fraction.
8. Process in accordance with Claims 1 to 7, characterized in that the fractions obtained in each condensation step are undercooled before they are expanded.
9. Process in accordance with Claims 1 through 8, characterized in that the compression of the multicomponent mixture takes place in two steps, that the multicomponent mixture obtained after the first compression step is partially condensed and subjected to phase separation, and that the liquid fraction obtained during phase separation is undercooled and expanded, while the gaseous fraction obtained during phase separation is conveyed directly to the second compression step.



10. Process in accordance with Claim 9, characterized in that the gaseous fraction obtained during partial condensation is partially condensed in heat exchange with the expanded liquid fraction and that the liquid fraction obtained in this process is undercooled and expanded, while the gaseous fraction obtained in this process is conveyed to the second compression step directly.

11. Process in accordance with Claims 1 to 10, characterized in that the multicomponent mixture consists of hydrocarbons with one, two, three, four, and possibly five or more carbon atoms.





